

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner  
 US Department of Commerce  
 United States Patent and Trademark  
 Office, PCT  
 2011 South Clark Place Room  
 CP2/5C24  
 Arlington, VA 22202  
 ETATS-UNIS D'AMERIQUE  
 in its capacity as elected Office

<b>Date of mailing (day/month/year)</b> 30 March 2001 (30.03.01)	
<b>International application No.</b> PCT/EP00/06843	<b>Applicant's or agent's file reference</b> PD-6517
<b>International filing date (day/month/year)</b> 18 July 2000 (18.07.00)	<b>Priority date (day/month/year)</b> 22 July 1999 (22.07.99)
<b>Applicant</b> SCOTT, Robert et al	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
 27 January 2001 (27.01.01)

☐ in a notice effecting later election filed with the International Bureau on:  
 \_\_\_\_\_

2. The election ☒ was

☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland  Facsimile No.: (41-22) 740.14.35	Authorized officer  Claudio Borton  Telephone No.: (41-22) 338.83.38
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The demand must be filed directly with the competent International Preliminary Examining Authority, or, if two or more Authorities are competent, with the one chosen by the applicant. The full name or two-letter code of that Authority may be indicated by the applicant on the line below:  
**IPEA/ EPO**

# PCT

## CHAPTER II

### DEMAND

under Article 31 of the Patent Cooperation Treaty:  
 The undersigned requests that the international application specified below be the subject of international preliminary examination according to the Patent Cooperation Treaty and hereby elects all eligible States (except where otherwise indicated).

For International Preliminary Examining Authority use only	
Identification of IPEA	Date of receipt of DEMAND
<b>Box No. I IDENTIFICATION OF THE INTERNATIONAL APPLICATION</b>	
Applicant's or agent's file reference PD-6517-11-CWA	
International application No. PCT/EP00/06843	International filing date (day/month/year) 18/July/2000
(Earliest) Priority date (day/month/year) 22/July/1999	
Title of invention PULLULAN FILM COMPOSITIONS	
<b>Box No. II APPLICANT(S)</b>	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)	
WARNER-LAMBERT COMPANY 201 Tabor Road Morris Plains, New Jersey 07950 US	
Telephone No.: ++973-540-2577	
Facsimile No.: ++973-540-3117	
Teleprinter No.:	
State (that is, country) of nationality: US	State (that is, country) of residence: US
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)	
SCOTT, Robert Königin Elisabethplein 26 bus 4 B-9100 Sint Niklaas Belgium	
State (that is, country) of nationality: GB	State (that is, country) of residence: BE
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)	
CADE, Dominique 11, rue des Américains F-68000 Colmar France	
State (that is, country) of nationality: FR	State (that is, country) of residence: FR
<input checked="" type="checkbox"/> Further applicants are indicated on a continuation sheet.	

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## Continuation of Box N . II APPLICANT(S)

*If none of the following sub-boxes is used, this sheet should not be included in the demand.*

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

HE, Xiongwei  
3, rue du Jura  
F-68280 Andolsheim  
France

State (that is, country) of nationality:  
China

State (that is, country) of residence:  
FR

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

State (that is, country) of nationality:

State (that is, country) of residence:

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

State (that is, country) of nationality:

State (that is, country) of residence:

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

State (that is, country) of nationality:

State (that is, country) of residence:

☐

Further applicants are indicated on another continuation sheet.

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**Box No. III AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE**The following person is ☒ agent ☐ common representativeand ☐ has been appointed earlier and represents the applicant(s) also for international preliminary examination.☐ is hereby appointed and any earlier appointment of (an) agent(s)/common representative is hereby revoked.☒ is hereby appointed, specifically for the procedure before the International Preliminary Examining Authority, in addition to the agent(s)/common representative appointed earlier.Name and address: *(Family name followed by given name; for a legal entity, full official designation.  
The address must include postal code and name of country.)*Ivo Mansmann  
Dr. Rudolf Tesch  
Warner-Lambert Company  
Legal Division, Patent Department  
c/o Gödecke GmbH  
Mooswaldallee 1  
79090 Freiburg, Germany

Telephone No.:

(0761)518-2525

Facsimile No.:

(0761)518-3076

Teleprinter No.:

☐ Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.**Box No. IV BASIS FOR INTERNATIONAL PRELIMINARY EXAMINATION****Statement concerning amendments:\***

1. The applicant wishes the international preliminary examination to start on the basis of:

☒ the international application as originally filedthe description ☐ as originally filed  
☐ as amended under Article 34the claims ☐ as originally filed  
☐ as amended under Article 19 (together with any accompanying statement)  
☐ as amended under Article 34the drawings ☐ as originally filed  
☐ as amended under Article 342. ☐ The applicant wishes any amendment to the claims under Article 19 to be considered as reversed.3. ☐ The applicant wishes the start of the international preliminary examination to be postponed until the expiration of 20 months from the priority date unless the International Preliminary Examining Authority receives a copy of any amendments made under Article 19 or a notice from the applicant that he does not wish to make such amendments (Rule 69.1(d)). *(This check-box may be marked only where the time limit under Article 19 has not yet expired.)*

\* Where no check-box is marked, international preliminary examination will start on the basis of the international application as originally filed or, where a copy of amendments to the claims under Article 19 and/or amendments of the international application under Article 34 are received by the International Preliminary Examining Authority before it has begun to draw up a written opinion or the international preliminary examination report, as so amended.

Language for the purposes of international preliminary examination: .....

☒ which is the language in which the international application was filed.☐ which is the language of a translation furnished for the purposes of international search.☐ which is the language of publication of the international application.☐ which is the language of the translation (to be) furnished for the purposes of international preliminary examination.**Box No. V ELECTION OF STATES**The applicant hereby elects all eligible States *(that is, all States which have been designated and which are bound by Chapter II of the PCT)*

excluding the following States which the applicant wishes not to elect:

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**Box No. VI CHECK LIST**

The demand is accompanied by the following elements, in the language referred to in Box No. IV, for the purposes of international preliminary examination:

- |  |   |        |
|--|---|--------|
| 1. translation of international application                              | : | sheets |
| 2. amendments under Article 34   | : | sheets |
| 3. copy (or, where required, translation) of amendments under Article 19 | : | sheets |
| 4. copy (or, where required, translation) of statement under Article 19  | : | sheets |
| 5. letter  | : | sheets |
| 6. other ( <i>specify</i> )  | : | sheets |

For International Preliminary Examining Authority use only

received                      not received

<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	<input type="checkbox"/>

The demand is also accompanied by the item(s) marked below:

- |   |   |
|---|---|
| 1. <input checked="" type="checkbox"/> fee calculation sheet  | 4. <input type="checkbox"/> statement explaining lack of signature                                  |
| 2. <input type="checkbox"/> separate signed power of attorney                                       | 5. <input type="checkbox"/> nucleotide and or amino acid sequence listing in computer readable form |
| 3. <input checked="" type="checkbox"/> copy of general power of attorney; reference number, if any. | 6. <input type="checkbox"/> other ( <i>specify</i> ):   |

**Box No. VII SIGNATURE OF APPLICANT, AGENT OR COMMON REPRESENTATIVE**

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the demand).



Dr. R. Tesch  
European Patent Attorney  
General Authorisation No. 29754  
January 24, 2001

For International Preliminary Examining Authority use only

1. Date of actual receipt of DEMAND:

2. Adjusted date of receipt of demand due to CORRECTIONS under Rule 60.1(b):

- |  |   |
|--|---|
| 3. <input type="checkbox"/> The date of receipt of the demand is AFTER the expiration of 19 months from the priority date and item 4 or 5, below, does not apply.                        | <input type="checkbox"/> The applicant has been informed accordingly. |
| 4. <input type="checkbox"/> The date of receipt of the demand is WITHIN the period of 19 months from the priority date as extended by virtue of Rule 80.5.                               |   |
| 5. <input type="checkbox"/> Although the date of receipt of the demand is after the expiration of 19 months from the priority date, the delay in arrival is EXCUSED pursuant to Rule 82. |   |

For International Bureau use only

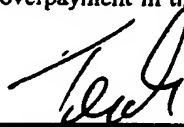
Demand received from IPEA on:

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## PCT

## FEE CALCULATION SHEET

## Annex to the Demand for international preliminary examination

<table border="1" style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%;">International application No.</td> <td style="width: 50%;">PCT/EP00/06843</td> </tr> <tr> <td>Applicant's or agent's file reference</td> <td>PD-6517-11-CWA</td> </tr> </table>	International application No.	PCT/EP00/06843	Applicant's or agent's file reference	PD-6517-11-CWA	<div style="border: 1px solid black; padding: 5px;"> For International Preliminary Examining Authority use only </div> <div style="border: 1px solid black; padding: 5px; margin-top: 10px;"> Date stamp of the IPEA </div>																				
International application No.	PCT/EP00/06843																								
Applicant's or agent's file reference	PD-6517-11-CWA																								
Applicant Warner-Lambert Company 201 Tabor Road Morris Plains, New Jersey 07950, US																									
<b>Calculation of prescribed fees</b>  <table style="width: 100%;"> <tr> <td style="width: 60%;">1. Preliminary examination fee .....</td> <td style="width: 20%; text-align: center;">EUR</td> <td style="width: 15%; text-align: center;">1.533,00</td> <td style="width: 5%; text-align: center;">P</td> </tr> <tr> <td colspan="4" style="height: 20px;"></td> </tr> <tr> <td>2. Handling fee <i>(Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.)</i> .....</td> <td style="text-align: center;">EUR</td> <td style="text-align: center;">147,00</td> <td style="text-align: center;">H</td> </tr> <tr> <td colspan="4" style="height: 20px;"></td> </tr> <tr> <td>3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box .....</td> <td style="text-align: center;">EUR</td> <td style="text-align: center;">1.680,00</td> <td></td> </tr> <tr> <td colspan="2"></td> <td colspan="2" style="text-align: center; border: 1px solid black;">TOTAL</td> </tr> </table>		1. Preliminary examination fee .....	EUR	1.533,00	P					2. Handling fee <i>(Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.)</i> .....	EUR	147,00	H					3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box .....	EUR	1.680,00				TOTAL	
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2. Handling fee <i>(Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.)</i> .....	EUR	147,00	H																						
3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box .....	EUR	1.680,00																							
		TOTAL																							
<b>Mode of Payment</b>  <table style="width: 100%;"> <tr> <td><input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)</td> <td><input type="checkbox"/> cash</td> </tr> <tr> <td><input type="checkbox"/> cheque</td> <td><input type="checkbox"/> revenue stamps</td> </tr> <tr> <td><input type="checkbox"/> postal money order</td> <td><input type="checkbox"/> coupons</td> </tr> <tr> <td><input type="checkbox"/> bank draft</td> <td><input type="checkbox"/> other (specify):</td> </tr> </table>		<input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)	<input type="checkbox"/> cash	<input type="checkbox"/> cheque	<input type="checkbox"/> revenue stamps	<input type="checkbox"/> postal money order	<input type="checkbox"/> coupons	<input type="checkbox"/> bank draft	<input type="checkbox"/> other (specify):																
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<input type="checkbox"/> postal money order	<input type="checkbox"/> coupons																								
<input type="checkbox"/> bank draft	<input type="checkbox"/> other (specify):																								
<b>Deposit Account Authorization</b> <i>(this mode of payment may not be available at all IPEAs)</i>  The IPEA/ EPO <input checked="" type="checkbox"/> is hereby authorized to charge the total fees indicated above to my deposit account.  <input checked="" type="checkbox"/> <i>(this check-box may be marked only if the conditions for deposit accounts of the IPEA so permit)</i> is hereby authorized to charge any deficiency or credit any overpayment in the total fees indicated above to my deposit account.																									
2800 0553 Deposit Account Number	24/January/2001 Date (day/month/year)	 Signature																							

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From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

MANSMANN, Ivo  
Warner-Lambert Company  
Legal Division  
c/o Gödecke AG, Patents  
Mooswaldallee 1  
D-79090 Freiburg  
ALLEMAGNE

PCT

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT

(PCT Rule 71.1)

Patentwesen

Eing.: 22. Okt. 2001

Date of mailing  
(day/month/year)

18.10.2001

Applicant's or agent's file reference

PD-6517-11-CWA

IMPORTANT NOTIFICATION

International application No.

PCT/EP00/06843

International filing date (day/month/year)

18/07/2000

Priority date (day/month/year)

22/07/1999

Applicant

WARNER-LAMBERT COMPANY

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



European Patent Office - P.B. 5818 Patentlaan 2  
NL-2280 HV Rijswijk - Pays Bas  
Tel. +31 70 340 - 2040 Tx: 31 651 epo nl  
Fax: +31 70 340 - 3016

Authorized officer

Sinanovic, E

Tel. +31 70 340-2672



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# PATENT COOPERATION TREATY

# PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>PD-6517-11-CWA</b>	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/EP00/06843</b>	International filing date (day/month/year) <b>18/07/2000</b>	Priority date (day/month/year) <b>22/07/1999</b>
International Patent Classification (IPC) or national classification and IPC <b>C08J5/18</b>		
Applicant <b>WARNER-LAMBERT COMPANY</b>		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 6 sheets, including this cover sheet.
 

☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of      sheets.

3. This report contains indications relating to the following items:
 

I	<input checked="" type="checkbox"/>	Basis of the report
II	<input type="checkbox"/>	Priority
III	<input type="checkbox"/>	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
IV	<input type="checkbox"/>	Lack of unity of invention
V	<input checked="" type="checkbox"/>	Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
VI	<input type="checkbox"/>	Certain documents cited
VII	<input type="checkbox"/>	Certain defects in the international application
VIII	<input type="checkbox"/>	Certain observations on the international application

Date of submission of the demand  <b>27/01/2001</b>	Date of completion of this report  <b>18.10.2001</b>
Name and mailing address of the international preliminary examining authority:  <div style="display: flex; align-items: center;"> <div>             European Patent Office - P.B. 5818 Patentlaan 2              NL-2280 HV Rijswijk - Pays Bas              Tel. +31 70 340 - 2040 Tx: 31 651 epo nl              Fax: +31 70 340 - 3016           </div> </div>	Authorized officer  <b>Lensen, H</b>  Telephone No. +31 70 340 2428



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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP00/06843

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, pages:**

1-18 as originally filed

**Claims, No.:**

1-41 as originally filed

**Drawings, sheets:**

1/1 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).  
☐ the language of publication of the international application (under Rule 48.3(b)).  
☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.  
☐ filed together with the international application in computer readable form.  
☐ furnished subsequently to this Authority in written form.  
☐ furnished subsequently to this Authority in computer readable form.  
☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.  
☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP00/06843

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes:	Claims	3,10,29-30,33-34,38-41
	No:	Claims	1-2,4-9,11-28,31-32,35-37
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-41
Industrial applicability (IA)	Yes:	Claims	1-41
	No:	Claims	

2. Citations and explanations  
**see separate sheet**

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP00/06843

**Re Item V**

**Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1). The following documents (D1-D5) are referred to in this communication; the numbering will be adhered to in the rest of the procedure:

D1 : US-A-4623394

D2 : JP-A-5065222

=D2A : PAJ Vol. 17, 376 (C1084)

=D2B : Derwent/WPI 1993-129084 [16]

D3 : JP-A-53079972

=D3A : WPI/Derwent 1978-59705A [33]

=D3B : C.A. Vol. 89, no. 18, 30/10/1978, abstract no. 147859

D4 : EP-A-328317

D5 : JP-A-4363332

=D5A : PAJ Vol. 17, no. 232 (C-1056)

=D5B : WPI/Derwent 1993-039394 [05]

2). Art. 33(2) PCT (Novelty) :

D1 discloses moulded articles such as films or capsules comprising a blend of a heteromannan and pullulan.

The heteromannans usable include galactomannans such as guar gum, tara gum, locust bean gum and glucomannans such as konjak gum, and are present in an amount of 1 to 80% by weight of the pullulan.

One or more substances such as colouring agent, pharmaceutical or tasting agent, can be incorporated into the moulded articles.

The film product formed thereof is more resistant to moisture and water, is superior in mechanical properties, is high in gas-barrier abilities and very low in oxygen-, air-, and flavour permeabilities. In experiment 1, mixtures are prepared containing pullulan and polysaccharide such as carrageenan. Example 1 discloses a film composition comprising pullulan and guar gum. A hard capsule is prepared in example 3 from an 15% aqueous pullulan solution containing 20% guar gum and 1% maltitol. The present application does not meet the requirements of Article 33(2) PCT, because the subject-

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP00/06843

matter of claims 1-2, 4-9 and 13-28 appears to be not new.

D2 discloses the manufacture of a film containing pullulan blended with gelatin, agar or carrageenan in an amount of about 5-10% based on the total weight of the film. The capsule may be soft or hard (see especially D2B).

In view of the disclosure of the abstracts D2A and D2B, the present application does not meet the requirements of Article 33(2) PCT, because the subject-matter of claims 1-2, 6, 17-19, 22, 25 and 26 appears to be not new.

D3 discloses the manufacture of a heat sealing film or sheet. The sheet may contain water-soluble substances compatible with pullulan such as starch (see D3A). The films are coated with water on the area to be sealed with water and heat-sealed at  $< 110^{\circ}\text{C}$ . In view of the disclosure of D3A and D3B, the subject-matter of claims 1, 6-8, 12 and 31-32 of the present application appears to be not novel.

D4 discloses the manufacture of edible films comprising curdlan and pullulan (see example 3). It has a feeling of transparency and is heat sealable.

The film is prepared by mixing the two elements in water and the aqueous mixture is extruded in the form of a thin film evenly onto a polyester film and heated for gelation. This thin film gel is passed through hot air, dried and finally peeled off the polyester film and wound up. The films may be used as wrapping films for foodstuffs (see page 3, lines 47-55).

The subject-matter of claims 1, 6-8, 13, 23 and 31 appears to be not novel.

D5 discloses moulded products made from a polysaccharide such as pullulan and 5-50 wt% water. The composition also comprises an inorganic filler such as  $\text{TiO}_2$  and/or a bulk filler such as starch. The moulded part is used as films, sheets, capsules, cups and bottles.

The subject-matter of claims 1, 6-9, 11-12, 19, 23, 31 and 35-37 appears to be not novel.

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP00/06843

3). Art. 33(3) PCT (Inventive step) :

The subject-matter of the claims 3, 10, 29-30, 33-34 and 38-41 does not involve an inventive step since they are dealing with for the skilled person obvious technical variations and cannot, when brought into the main claim, establish an inventive step.

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# PATENT COÖPERATION TREATY

- Mitteilung an Erfinder

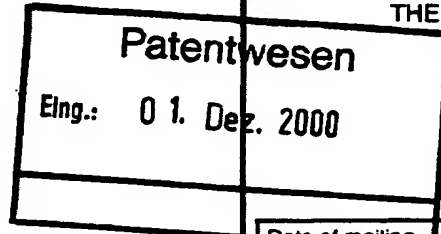
6517  
PCT

From the INTERNATIONAL SEARCHING AUTHORITY

## PCT

To:

GÖDECKE AG,  
PATENTS  
Attn. TESCH, Rudolf  
Mooswaldallee 1  
D-79090 Freiburg  
GERMANY



NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL SEARCH REPORT  
OR THE DECLARATION

(PCT Rule 44.1)

Date of mailing  
(day/month/year)

01/12/2000

Applicant's or agent's file reference

PD-6517

**FOR FURTHER ACTION**

See paragraphs 1 and 4 below

International application No.

PCT/EP 00/06843

International filing date

(day/month/year)

18/07/2000

Applicant

WARNER-LAMBERT COMPANY

1. ☒ The applicant is hereby notified that the International Search Report has been established and is transmitted herewith.

**Filing of amendments and statement under Article 19:**

The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 46):

**When?** The time limit for filing such amendments is normally 2 months from the date of transmittal of the International Search Report; however, for more details, see the notes on the accompanying sheet.

**Where?** Directly to the International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland  
Facsimile No.: (41-22) 740.14.35

For more detailed instructions, see the notes on the accompanying sheet.

2. ☐ The applicant is hereby notified that no International Search Report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.

3. ☐ With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

☐ the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.

☐ no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Further action(s):** The applicant is reminded of the following:

Shortly after 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.

Within 19 months from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).

Within 20 months from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.

Name and mailing address of the International Searching Authority



European Patent Office, P.B. 5818 Patentaan 2  
NL-2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Alfredo Prein

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## NOTES TO FORM PCT/ISA/220

These Notes are intended to give the basic instructions concerning the filing of amendments under article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article", "Rule", and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions respectively.

### INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international publication. Furthermore, it should be emphasized that provisional protection is available in some States only.

#### What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

#### When?

Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

#### Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been/is filed, see below.

#### How?

Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

#### What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

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The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]:  
"Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
2. [Where originally there were 15 claims and after amendment of all claims there are 11]:  
"Claims 1 to 15 replaced by amended claims 1 to 11."
3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:  
"Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or  
"Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
4. [Where various kinds of amendments are made]:  
"Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

**"Statement under article 19(1)" (Rule 46.4)**

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)".

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

**Consequence if a demand for international preliminary examination has already been filed**

If, at the time of filing any amendments under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the same time of filing the amendments with the International Bureau, also file a copy of such amendments with the International Preliminary Examining Authority (see Rule 62.2(a), first sentence).

**Consequence with regard to translation of the international application for entry into the national phase**

The applicant's attention is drawn to the fact that, where upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see Volume II of the PCT Applicant's Guide.

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# PATENT COOPERATION TREATY

# PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>PD-6517</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/EP 00/ 06843</b>	International filing date (day/month/year) <b>18/07/2000</b>	(Earliest) Priority Date (day/month/year) <b>22/07/1999</b>
Applicant  <b>WARNER-LAMBERT COMPANY</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

**1. Basis of the report**

a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.

☒ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

1

☐ None of the figures.

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/06843

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 7 C08J5/18 C08L5/00 A61K9/48

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08J C08L A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE WPI Week 197833 Derwent Publications Ltd., London, GB; AN 1978-59705A XP002152902 &amp; JP 53 079972 A (HAYASHIBARA BIOCHEMICAL LAB), 14 July 1978 (1978-07-14) abstract &amp; CHEMICAL ABSTRACTS, vol. 89, no. 18, 30 October 1978 (1978-10-30) Columbus, Ohio, US; abstract no. 147859, abstract</p>	1-41
X	<p>US 4 623 394 A (SATOSHI NAKAMURA ET AL.) 18 November 1986 (1986-11-18) example 3; table I</p>	1-30

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

\* Special categories of cited documents:

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*A\* document member of the same patent family

Date of the actual completion of the international search

15 November 2000

Date of mailing of the international search report

01/12/2000

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Lensen, H

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# INTERNATIONAL SEARCH REPORT

International Application No.

PCT/EP 00/06843

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 328 317 A (TAKEDA CHEMICAL INDUSTRIES) 16 August 1989 (1989-08-16) example 3 ---	1, 6, 13, 35-37
X	PATENT ABSTRACTS OF JAPAN vol. 17, no. 232 (C-1056), 12 May 1993 (1993-05-12) & JP 04 363332 A (MITSUBISHI RAYON) abstract & DATABASE WPI Week 199305 Derwent Publications Ltd., London, GB; AN 1993-039394 abstract ---	1
X	PATENT ABSTRACTS OF JAPAN vol. 17, no. 376 (C-1084), 15 July 1993 (1993-07-15) & JP 05 065222 A (FUJI CAPSULE), 19 March 1993 (1993-03-19) abstract & DATABASE WPI Week 199316 Derwent Publications Ltd., London, GB; AN 129084 abstract ---	1-30
A	US 3 784 390 A (HIROMI HIJIYA ET AL.) 8 January 1974 (1974-01-08) cited in the application -----	

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 00/06843

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
JP 53079972	A	14-07-1978	NONE	
US 4623394	A	18-11-1986	JP 1843452 C JP 5049705 B JP 60219238 A FR 2562899 A GB 2162528 A, B	12-05-1994 27-07-1993 01-11-1985 18-10-1985 05-02-1986
EP 328317	A	16-08-1989	CN 1036967 A JP 1289457 A	08-11-1989 21-11-1989
JP 04363332	A	16-12-1992	NONE	
JP 05065222	A	19-03-1993	NONE	
US 3784390	A	08-01-1974	JP 48051777 A JP 52006346 B AR 200845 A AR 200858 A AU 468995 B AU 4457672 A BE 786574 A CA 1007415 A CH 564574 A DE 2235991 A ES 405103 A FI 54133 B FR 2147112 A GB 1374199 A IT 1048891 B NL 7210152 A, B, NO 138662 B PH 9411 A SE 384517 B SU 591152 A ZA 7205017 A	20-07-1973 21-02-1977 27-12-1974 27-12-1974 29-01-1976 17-01-1974 16-11-1972 29-03-1977 31-07-1975 08-02-1973 16-11-1975 30-06-1978 09-03-1973 20-11-1974 20-12-1980 25-01-1973 10-07-1978 11-11-1975 10-05-1976 30-01-1978 25-04-1973

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(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
1 February 2001 (01.02.2001)

PCT

(10) International Publication Number  
**WO 01/07507 A1**

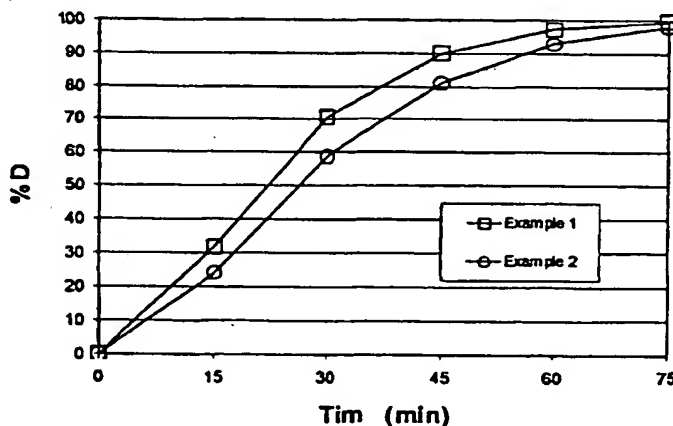
- (51) International Patent Classification<sup>7</sup>: C08J 5/18, des Americans, F-68000 Colmar (FR). HE, Xiongwei  
C08L 5/00, A61K 9/48 [CN/FR]; 3, rue du Jura, F-68280 Andolsheim (FR).
- (21) International Application Number: PCT/EP00/06843 (74) Agents: MANSMANN, Ivo et al.; Gödecke AG, Patents,  
Mooswaldallee 1, D-79090 Freiburg (DE).
- (22) International Filing Date: 18 July 2000 (18.07.2000)
- (25) Filing Language: English (81) Designated States (*national*): CA, CN, ID, JP, KR, MX, US.
- (26) Publication Language: English (84) Designated States (*regional*): Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).
- (30) Priority Data:  
99401849.7 22 July 1999 (22.07.1999) EP  
00401333.0 16 May 2000 (16.05.2000) EP
- (71) Applicant (*for all designated States except US*):  
WARNER-LAMBERT COMPANY [US/US]; 201  
Tabor Road, Morris Plains, NJ 07950 (US).
- (72) Inventors; and
- (75) Inventors/Applicants (*for US only*): SCOTT, Robert  
[GB/BE]; Königin Elisabethplein 26, bus 4, B-9100  
Sint Niklaas (BE). CADE, Dominique [FR/FR]; 11, rue

**Published:**

- With international search report.
- Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: PULLULAN FILM COMPOSITIONS



(57) Abstract: The invention concerns compositions based on pullulan and a setting system for the use in pharmaceutical, veterinary, food, cosmetic or other products like films for wrapping food, aspics or jellies, preferably for predosed formulations like soft or hard capsules. The composition preferably further contains a surfactant. By using aqueous solution of the inventive compositions, the hard pullulan capsules are produced by a conventional dipping moulding process under the same process condition range than conventional gelatine capsules.

PTO/PCT Rec'd 22 JAN 2002

WO 01/07507 A1

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## Pullulan Film Compositions

5 The invention concerns pullulan compositions for the use in pharmaceutical, veterinary, food, cosmetic or other products like films for wrapping food, aspics or jellies, preferably for predosed formulations like soft or hard capsules.

Conventional hard capsules are made with gelatine by dip moulding process. The dip molding process is based on the setting ability of hot gelatine solutions by cooling. For the industrial manufacture of pharmaceutical capsules gelatine is most preferred for its gelling, film forming and surface active properties. The manufacture of hard gelatine capsules by dip moulding process exploits fully its gelling and film forming abilities. A typical dip moulding process comprises the steps of dipping mould pins into a hot solution of gelatine, removing the pins from the gelatine solution, allowing the gelatine solution attached on pins to set by cooling, drying and stripping the so-formed shells from the pins. The setting of the solution on the mould pins after dipping is the critical step to obtain a uniform thickness of the capsule shell.

On a totally automatic industrial hard gelatine capsule machine, the process consists to dip mould pins into hot gelatine solution, to remove the pins from the solution, to turn the pins from downside to upside, to dry the gelatine solution (gel) attached on the pins, to strip the capsule shell and finally to cut and pre-joint the cap and body. The immediate setting of the gelatine solution on the dip pins after dipping is the key step in the process. Otherwise, the gelatine solution would flow down, leading

to a very low top thickness, and no capsule of quality could be produced.

Attempts have been made to manufacture capsules with materials other than gelatine, notably with modified cellulose. Successful industrial examples are the capsules made of hydroxypropyl methylcellulose (HPMC).

Pullulan is a natural, viscous polysaccharide extracellularly produced by growing certain yeasts on starch syrups. It has good film forming properties and a particularly low oxygen permeability. Its existence was reported for the first time in 1938. Hayashibara Company started the commercial production in 1976.

There are numerous patents about the use of pullulan in moulded articles, edible films, and coatings.

US 4,623,394 describes a molded article which exhibits a controlled desintegrability under hydrous conditions. The composition of the molded article consists essentially of a combination of pullulan and a heteromannan, the amount of heteromannan being, based on the dry solids, 1 to 100% of the pullulan.

JP5-65222-A describes a soft capsule, capable of stabilizing a readily oxidizable substance enclosed therein, exhibiting easy solubility, and being able to withstand a punching production method. The soft capsule is obtained by blending a capsule film substrate such as gelatin, agar, or carrageenan with pullulan.

US-3,784,390-A, corresponding to FR 2,147,112 and GB 1,374,199, discloses that certain mixtures of pullulan with at least one member of the group consisting of amylose, polyvinyl alcohol, and gelatine can be shaped by

compression molding or extrusion at elevated temperatures or by evaporation of water from its aqueous solutions to form shaped bodies, such as films or coatings. To retain the valuable properties of pullulan to an important extent the mixture should not contain more than 120 percent amylose, 100 percent polyvinyl alcohol, and/or 150 percent gelantine based on the weight of the pullulan in the mixture.

US 4,562,020, discloses a continuous process for producing a self-supporting glucan film, comprising casting an aqueous glucan solution on the surface of a corona-treated endless heat-resistant plastic belt, drying the glucan solution thereon while heating and releasing the resultant self-supporting glucan film. Suitable glucans are those which substantially consist of repeating maltotriose units, such as pullulan or elsinan.

JP-60084215-A2 discloses a film coating composition for a solid pharmaceutical having improved adhesive properties on the solid agent. The film is obtained by incorporating pullulan with a film coating base material such as methylcellulose.

JP-2000205-A2 discloses a perfume-containing coating for a soft capsule. The coating is obtained by adding a polyhydric alcohol to a pullulan solution containing an oily perfume and a surfactant such as a sugar ester having a high HLB.

US 2,949,397 describes a method of making a mineral filled paper which comprises the step of coating finely divided mineral filler particles with an aqueous colloidal dispersion of plant mucilage in the form of substituted mannan selected from the group consisting of manno-glactans and gluco-glactans.

US 3,871,892 describes the preparation of pullulan esters by the reaction of pullulan with aliphatic or aromatic fatty acids or their derivatives in the presence of suitable solvents and/or catalysers. The pullulan esters can be shaped by compression molding or extrusion at elevated temperatures or by evaporation of solvents from their solutions to form shaped bodies such as films or coatings.

US 3,873,333 discloses adhesives or pastes prepared by dissolving or dispersing uniformly a pullulan ester and/or ether in water or in a mixture of water and acetone in an amount between 5 percent to 40 percent of the solvent.

US 3,932,192 describes a paper coating material containing pullulan and a pigment.

US 4,257,816 discloses a novel blend of algin, TKP, and guar gum which are useful in commercial gum applications, particularly for the paper-industry, where thickening, suspending, emulsifying, stabilizing, film-forming and gel-forming are needed.

US 3,997,703 discloses a multilayered molded plastic having at least one layer comprising pullulan and at least one layer selected from the group consisting of layers composed of homopolymers and copolymers of olefins and/or vinyl compounds, polyesters, polyamides, celluloses, polyvinylalcohol, rubber hydrochlorides, paper, and aluminum foil.

GB 1,533,301 describes a method of improving the water-resistance of pullulan by the addition of a water-soluble dialdehyde polysaccharides to pullulan.

GB 1559 644 also describes a method of improving the water-resistance of pullulan articles. The improved articles are manufactured by means of a process comprising bringing a mixture or shaped composition of a (a) pullulan or a water soluble derivative thereof and (b) polyuronide or a water-soluble salt thereof in contact with an aqueous and/or alcoholic solution of a di- or polyvalent metallic ion.

Although capsules were mentioned or claimed in these patents, their compositions do not have sufficient setting ability or none at all. Consequently, these compositions do not allow an industrial scale hard capsule production, and no attempt has been described to produce pullulan hard capsules by means of conventional dipping moulding processes.

Another problem with conventional pullulan hard capsules is their poor surface gliding performance, which leads to a high opening force of the pre-joint capsules and a high closing force. Indeed, these are two key parameters for a good filling performance on automatic high speed capsule filling equipment. During the filling process, the filling equipment opens, fills and recloses the capsules in an extremely high cadence. High opening or closing force can lead to defects such as non open, punched capsule ends and etc, and consequently to frequent machine stops.

The object of the present invention is therefore the provision of improved pullulan compositions which overcome the drawbacks of the prior art compositions. This object is solved according to the film forming composition, the container for unit dosage, the caplets, the capsules, the aqueous solutions, the use of the aqueous solutions for the manufacturing of hard capsules in a dip molding process, and the manufacturing of hard capsules from aqueous pullulan solutions according to the independent claims.

Further advantageous features, aspects and details of the invention are evident from the dependent claims, the description and the drawings. The claims are to be understood as a first non-limiting approach to define the invention in general terms.

The invention provides a film-forming composition comprising pullulan and a setting system.

Surprisingly, we found that the addition of a very small amount of a setting system, preferably comprising hydrocolloids acting as a gelling agent, most preferably polysaccharides, confers an appropriate setting ability with cooling to pullulan solution so that the production of hard pullulan capsules can be produced with a conventional dip moulding process.

In a preferred embodiment, the film forming composition may preferably further contain a cation containing salt, comprising at least one cation. Optionally, the film forming composition may further comprise at least one sequestering agent.

In an aspect of the present invention the film compositions are used for the manufacturing of hard capsules by conventional dip moulding process as normally used in the production of conventional hard gelatine capsules.

In an additional aspect of the present invention there are provided aqueous solutions comprising the film forming compositions of the present invention for the manufacture of capsules. The setting system gets the solution to set on the dipped pins, thus assuring a uniform capsule shell thickness. The setting system is preferably composed of a gelling agent, such as said hydrocolloids or



polysaccharides, and optionally salt and sequestering agent.

The cation containing salt in the composition serves to enhance the setting ability of the gelling agents. Preferably, the salt comprises cations such as  $K^+$ ,  $Li^+$ ,  $Na^+$ ,  $NH_4^+$ ,  $Ca^{2+}$ , or  $Mg^{2+}$ , etc. The amount of cations is preferably less than 3%, especially 0.01 to 1% by weight in the aqueous pullulan solution. The preferred salt concentration in the solution is less than 2%.

In a further aspect of the present invention there are provided compositions for the use in pharmaceutical, veterinary, food, cosmetic or other products like films for wrapping food, aspics or jellies, preferably for predosed formulations like soft or hard capsules and wherein the pullulan compositions has in aqueous solution a sufficient setting ability.

In a particular aspect of the present invention there are provided containers for unit dosage forms for agrochemicals, seeds, herbs, foodstuffs, dyestuffs, pharmaceuticals, or flavoring agents produced from the film forming compositions of the present invention. Preferably, such containers are capsules, especially pharmaceutical capsules. The capsule halves of the capsules are preferably sealed with one or more layers of the film forming compositions of the present invention. The capsule halves are preferably sealed by means of a liquid fusion process. The capsules of the present invention may preferably release the product they are filled with at low temperatures, preferably at room temperature.

In a further aspect of the present invention there are provided caplets encapsulated in a film forming composition of the present invention.

Compared to gelatine or HPMC, the advantages of pullulan can be mentioned as follows:

- Non-animal origin
- No chemical modification, totally natural.
- 5 • Higher product quality consistency by the fermentation process control.
- High homogeneity and transparency of films
- Very low oxygen permeability. Its capsules are particularly useful for the filling of oxygen sensitive products such as fish and vegetable oils.
- 10 • Relatively low water content, lower than gelatine.
- High stability of various properties over storage such as mechanical and dissolution properties.

15 The addition of a setting system, preferably based on polysaccharides, to pullulan solutions enables the adaptation of specific and desired gelling properties for the production of hard pullulan capsules by a conventional dipping process. For the production of such capsules it is extremely important that the film forming pullulan solution  
20 remaining on the mould pins after dipping is prohibited from flowing down the pins. Otherwise the obtained film will not have the desired uniform thickness.

25 Consequently the present invention enables that the hard pullulan capsules can be produced with the same equipment used for the production of conventional hard gelatine capsules in the same range of process conditions. Furthermore capsules produced from compositions of the

present invention have the same dimensional specifications and allow the use of the existing filling machinery and do not require specific and new equipment for the filling process.

5 In an preferred embodiment of the present invention, the concentration of pullulan in the dipping aqueous solution is in a range of 10 to 60%, preferably 10 to 50%, more preferably 15 to 40%, and most preferably 10 to 40% by weight.

10 Although pullulan of various molecular weight is usable, pullulan has a viscosity from 100 cps to 2000 cps at above mentioned concentration and at dipping temperature (40-70°C) is preferred.

15 The pullulan without desalting (Japanese food grade) is usable, however the desalted pullulan (Japanese pharmaceutical excipients grade) is preferable for its improved mechanical properties.

20 In preferred embodiments of the present invention the setting system comprises a hydrocolloid or mixtures of hydrocolloids.

Suitable hydrocolloids or mixtures thereof for the present invention, producing synergistic properties, may be selected from the group comprising natural seaweeds, natural seed gums, natural plant exudates, natural fruit  
25 extracts, biosynthetic gums, gelatines, biosynthetic processed starch or cellulosic materials, preferred are the polysaccharides.

30 In a preferred embodiment of the present invention, the polysaccharides are selected from the group comprising alginates, agar gum, guar gum, locust bean gum (carob),

5 carrageenan, tara gum, gum arabic, ghatti gum, Khaya grandifolia gum, tragacanth gum, karaya gum, pectin, arabian (araban), xanthan, gellan, starch, Konjac mannan, galactomannan, funoran, and other exocellular polysaccharides. Preferred are exocellular polysaccharides.

10 Preferred exocellular polysaccharides for use in the present invention are selected from the group comprising xanthan, acetan, gellan, welan, rhamsan, furcelleran, succinoglycan, scleroglycan, schizophyllan, tamarind gum, curdlan, and dextran.

In a further preferred embodiment of the present invention the hydrocolloids of the setting system are kappa-carrageenan or gellan gum or combinations like xanthan with locust bean gum or xanthan with konjac mannan.

15 Among the setting systems mentioned above, the systems of kappa-carrageenan with cations and gellan gum with cations are specifically preferred. They produce high gel strength at low concentrations and have good compatibility with pullulan.

20 The amount of the setting agent is preferably in the range of 0.01 to 5% by weight and especially preferred 0.03 to 1.0% in the aqueous pullulan solution of the present invention.

25 In a further preferred embodiment of the present invention the sequestering agents are selected from the group comprising ethylenediaminetetraacetic acid, acetic acid, boric acid, citric acid, edetic acid, gluconic acid, lactic acid, phosphoric acid, tartaric acid, or salts thereof, methaphosphates, dihydroxyethylglycine, lecithin or beta cyclodextrin and combinations thereof. Especially preferred  
30

is ethylenediaminetetraacetic acid or salts thereof or citric acid or salts thereof.

5 In another preferred embodiment of the present invention, the amount of the sequestering agent is preferably less than 3%, especially 0.01 to 1% by weight of the aqueous dipping solution.

10 In the case that gellan is used as gelling agent, the compositions preferably contain a sequestering agent to improve the capsule solubility. The preferred sequestering agents are ethylenediaminetetraacetic acid or salts thereof and citric acid and salts thereof. The amount is preferably less than 1% in the solution compositions.

15 The pullulan compositions of the present invention may in a further preferred embodiment additionally comprise pharmaceutically or food acceptable colouring agents in the range of from 0% to 10% based upon the weight of the film. The colouring agents may be selected from the group comprising azo-, quinophthalone-, triphenylmethane-, xanthene- or indigoid dyes, iron oxides or hydroxides, 20 titanium dioxide or natural dyes or mixtures thereof. Examples are patent blue V, acid brilliant green BS, red 2G, azorubine, ponceau 4R, amaranth, D+C red 33, D+C red 22, D+C red 26, D+C red 28, D+C yellow 10, yellow 2 G, FD+C yellow 5, FD+C yellow 6, FD+C red 3, FD+C red 40, FD+C blue 25 1, FD+C blue 2, FD+C green 3, brilliant black BN, carbon black, iron oxide black, iron oxide red, iron oxide yellow, titanium dioxide, riboflavin, carotenes, anthocyanines, turmeric, cochineal extract, chlorophyllin, canthaxanthin, caramel, or betanin.

30 The inventive pullulan compositions may in a further preferred embodiment additionally contain at least one

pharmaceutically or food acceptable plasticiser or flavoring agent.

5 In yet another preferred embodiment of the present invention the pullulan containers, such as capsules may be coated with a suitable coating agent like cellulose acetate phthalate, polyvinyl acetate phthalate, methacrylic acid gelatines, hypromellose phthalate, hydroxypropylmethyl cellulose phthalate, hydroxyalkyl methyl cellulose phthalates, hydroxypropyl methylcellulose acetate succinate  
10 or mixtures thereof to provide e.g. enteric properties.

In a preferred embodiment of the present invention, the film-forming compositions further comprise one or more surfactants.

15 The surfactant in the compositions improves the capsule surface properties in such a way that the capsule works well on the conventional automatic high speed capsule filling equipment.

20 We have surprisingly found that the addition of a small quantity of selected surfactants of pharmaceutical or food quality, we can improve dramatically the pullulan film surface gliding performance, consequently to get the capsule opening and closing forces to the range required by filling equipment.

25 Therefore, the present invention provides compositions for hard pullulan capsules with improved surface properties containing pullulan, setting system and surfactant and the aqueous solutions of said film forming compositions for the manufacturing of the capsules.

30 With these preferably aqueous solutions, we can produce hard pullulan capsules with good filling performance by

conventional dip mould process just like hard gelatine capsules.

5 A further perceived disadvantage of unmodified pullulan capsule film is its adhesive nature or tackiness when touched by hand. The rapid remoisturing properties of pullulan results in a perceived tackiness when holding the capsule film in the hand for 30 seconds or more.

10 An additional disadvantage is evident on swallowing the capsule film as the film may adhere to the tongue, palet (upper mouth), throat or oesophagus and compare unfavorably with traditional gelatin film capsules.

15 Patient compliance is a major advantage of the traditional hard gelatin capsule and supported by several market studies whcih cite "ease of swallow" as an important factor in patient preference for the capsule oral dosage form.

20 In order to solve this perceived disadvantage of pullulan capsule film, surprisingly it has been found that a surfactant content in the pullulan capsule film provides an acceptable temporary water-repellant surface for handling or swallowing the capsule. Additionally, the selected surfactant may be applied externally as a transparent coating in the range 0.5 to 100 microns. The selected surfactants are water soluble at 37C.

25 The pullulan in the compositions is the base material for hard capsule making. Its preferred concentration in the aqueous solutions comprising the surfactant is from 10 to 40%.

30 The preferred gelling agents for the use with the surfactant are kappa-carrageenan and gellan with a concentration in the solutions 0.05-3%.

The surfactant in the compositions is aimed to improve the capsule surface gliding performance, and so the capsule filling performance on filling equipment. The surfactant can be cationic, anionic, non-ionic or amphoteric, and preferably selected from pharmaceutical and food quality such as sodium lauryl sulphate (SLS), dioctyl sodium sulfosuccinate (DSS), benzalkonium chloride, benzethonium chloride, cetrimide (trimethyltetradecylammonium bromide), fatty acid sugar esters, glyceryl monooleate, polyoxyethylene sorbitan fatty acid esters, polyvinyl alcohol, dimethylpolysiloxan, sorbitan esters or lecithin. Its amount based on pullulan is preferably 0.01% to 3%.

The above mentioned and other features of the present invention will be better understood by reference to the following examples and the accompanying figure, in which:

Fig. 1 shows a graph listing dissolution test results of capsules according to the present invention filled with acetaminophen in deionized water at 37°C (USP XXIII dissolution).

The following examples and tests, not limitative, demonstrate the pullulan capsule production and properties. Furthermore, the examples demonstrate the hard capsule making, the surface gliding improvement, and the capsule filling improvement.

#### Example 1:

1.0 kg of pullulan (PI-20, Japanese Pharmaceutical Excipients grade) powder is mixed with 10 g of kappa-carrageenan. To 4.0 kg of deionised water under stirring at room temperature is added 20 g of potassium acetate (0.2% by weight in the solution), followed by addition of the above mixture (20% of pullulan and 0.2% of carrageenan in



the solution). The powder addition and stirring speeds should be very high in order to avoid the forming of lumps, which take a long time to be dissolved. The solution is heated up to 70°C under stirring to totally dissolve the carrageenan and pullulan. It is possible to dissolve the components directly at 70°C, but the tendency of pullulan to lump is much higher.

The pullulan solution thus prepared is defoamed under slow stirring and then poured into a dipping dish of a pilot machine of conventional hard gelatine capsule production equipment. While keeping the dipping pullulan solution at 60°C, natural transparent hard pullulan capsules of size 0 were produced according to the conventional process with the same dimensional specifications to the conventional hard gelatine capsules.

#### Example 2:

1.0 kg of pullulan (PI-20) powder is mixed with 6 g of gellan. To 4.0 kg of deionised water under stirring at room temperature is added 20 g of potassium acetate (0.4% by weight in the solution) and 2 g of ethylenediaminetetraacetic acid disodium salt (0.04% in the solution), followed by addition of the above mixture (20% of pullulan and 0.12% of gellan in the solution). Heat the solution up to 75°C under stirring to dissolve totally the gellan and pullulan.

The pullulan solution thus prepared is defoamed under slow stirring and then poured into a dipping dish of a pilot machine of conventional hard gelatine capsule production equipment. While keeping the dipping pullulan solution at 60°C, natural transparent hard capsules of size 0 were produced according to the conventional process with the

same dimensional specifications to the conventional hard gelatine capsules.

Disintegration test results:

Table 1: Disintegration test results (according to USP XXIII 1995-<701> Disintegration):

Capsule	Example 1	Example 2
Capsule emptied time	3.0 min	2.0 min
Total disintegration time	10.0 min	11.8 min

Dissolution test results of capsules filled with acetaminophen in deionised water at 37°C (USP XXIII dissolution) are represented in Fig. 1.

#### Example 3: Pullulan film gliding improvement

In 400g of demineralised water at room temperature were dispersed under stirring 0.05 g SLS (500ppm/pullulan), 1 g of kappa-carrageenan (0.2%), 1.25 g of potassium acetate (0.25%) and 100 g of pullulan (20%). The mixture is heated to 70°C under stirring for complete solubilisation and then the stirring is reduced for defoaming. The solution then is used to cast on glass plates of 4 mm thickness to form pullulan films of about 100 µm thickness after drying at room conditions.

The pullulan film gliding performance is evaluated by a test on a slanted plan, a method commonly used by gelatine producers. The method determines the smallest angle of inclination of glass plate to provoke the gliding of a film coated glass plate over another one with films face to

face. Consequently, the smaller the gliding angle, the better the film gliding performance.

Repeat the above example with surfactant contents listed in Table 2.

5 In Table 2, we gathered the gliding performance for different surfactants and quantity.

Table 2: Pullulan gliding performance (°)

Surfactant	No	500 ppm	1000 ppm	5000 ppm
SLS	29	9	5	6
Hydrolysed deoil lecithin		9	9	7
Polysorbate 20		12	12	
Polysorbate 80		10	9	
Sorbitan laurate		10	8	
Sorbitan oleate		9	7	

#### Example 4: Pullulan capsule production and performance

10 In 142 liters of demineralised water at room temperature were dispersed under stirring 20 g hydrolysed deoil lecithin (500ppm/pullulan), 363 g kappa-carrageenan (0.2%) and 40 kg pullulan (22%). The mixture is heated up to 70°C under stirring for total solubilisation. 455 g potassium acetate previously dissolved in some water was then added  
 15 into the solution. A slurry made with 800 g TiO<sub>2</sub>, 3 litres hot water and 3 liters so prepared pullulan solution by high shearing was added into the solution in order to produce white opaque capsules. After defoaming, the  
 20 solution is finally stabled at 60°C.

5 A second identical preparation was made. The two preparations were used to feed a conventional hard gelatine capsule production machine, white opaque hard pullulan capsules were then produced in the similar way to hard gelatine capsules.

As reference, transparent pullulan hard capsules without surfactant in the formulation were produced in the same way as above.

10 The improvement of hard pullulan capsules by the addition of surfactant is illustrated by the data gathered in Table 3, and was confirmed by a filling trial on a filling equipment KGF400.

Table 3

Capsule	Opening force of pre-lock capsule	Closing force
Capsule of example 2	12 g	6.0 N
Reference capsule	36 g	7.6 N

Claims

1. A film forming composition comprising pullulan and a setting system.
- 5 2. The film forming composition according to claim 1, wherein the setting system further comprises cations.
3. The film forming composition according to claim 2, wherein the cations are preferably selected from the group comprising  $K^+$ ,  $Na^+$ ,  $Li^+$ ,  $NH_4^+$ ,  $Ca^{++}$  and  $Mg^{++}$ .
- 10 4. The film forming compositions according to claims 1 or 2, wherein the setting system further comprises at least one sequestering agent.
- 15 5. The film forming composition according to claim 4, wherein the at least one sequestering agent is selected from the group comprising ethylenediaminetetraacetic acid, acetic acid, boric acid, citric acid, edetic acid, gluconic acid, lactic acid, phosphoric acid, tartaric acid, or salts thereof, methaphosphates, dihydroxyethylglycine, lecithin or beta cyclodextrin.
- 20 6. The film forming composition according to any one of claims 1 to 5, wherein the setting system comprises hydrocolloids.
7. The film forming composition according to claim 6, wherein the hydrocolloids of the setting system are selected from polysaccharides.
- 25 8. The film forming composition according to one of claims 6 or 7, wherein the hydrocolloids of the setting system are selected from exocellular polysaccharides.

9. The film forming composition according to any one of the preceding claims, wherein the content of pullulan is 85% to 95% by weight, and wherein the content of water is 5% to 15% by weight.
- 5 10. The film forming composition according to any one of the preceding claims, wherein the content of the cations is less than 5% by weight, preferably 0.01% to 3% by weight, more preferably 0.5% to 2% by weight.
- 10 11. The film forming composition according to any one of the preceding claims, wherein the content of the sequestering agent is less than 5% by weight, preferably 0.01% to 3% by weight, more preferably 0.5% to 2% by weight.
- 15 12. The film forming composition according to any one of claims 6 to 11, wherein the hydrocolloids of the setting system are selected from the group comprising alginates, agar gum, guar gum, locust bean gum (carob), carrageenan, tara gum, gum arabic, ghatti gum, Khaya grandifolia gum, tragacanth gum, karaya gum, pectin, 20 arabian (araban), xanthan, gellan, starch, Konjac mannan, galactomannan, or funoran.
- 25 13. The film forming composition according to any one of claims 6 to 11, wherein the hydrocolloids of the setting system are selected from the group comprising xanthan, acetan, gellan, welan, rhamsan, furcelleran, succinoglycan, scleroglycan, schizophyllan, tamarind gum, curdlan, or dextran.
- 30 14. The film forming composition according to any one of claims 6 to 11, wherein the hydrocolloids of the setting system are selected from gellan gum or kappa-carrageenan.

15. The film forming composition according to any one of claims 1 to 14, further containing plasticizers or/and flavoring agents.
- 5 16. The film forming composition according to claims 1 to 15, further containing colouring agents in a range from about 0% to 10% based upon the weight of the composition.
- 10 17. The film forming composition according to claim 16 wherein the colouring agent or mixture of colouring agents is selected from the group comprising azo-, quinophthalone-, triphenylmethane-, xanthene- or indigoid dyes, iron oxides or hydroxides, titanium dioxide or natural dyes.
- 15 18. The film forming composition according to claim 17 wherein the colouring agent or mixture of colouring agents is selected from the group comprising patent blue V, acid brilliant green BS, red 2G, azorubine, ponceau 4R, amaranth, D+C red 33, D+C red 22, D+C red 26, D+C red 28, D+C yellow 10, yellow 2 G, FD+C yellow 20 5, FD+C yellow 6, FD+C red 3, FD+C red 40, FD+C blue 1, FD+C blue 2, FD+C green 3, or brilliant black BN.
- 25 19. The film forming composition according to claim 16 wherein the colouring agent or mixture of colouring agents is selected from the group comprising carbon black, iron oxide black, iron oxide red, iron oxide yellow, titanium dioxide, riboflavin, carotenes, anthocyanines, turmeric, cochineal extract, chlorophyllin, canthaxanthin, caramel, or betanin.
- 30 20. The film forming composition according to any one of the preceding claims, wherein the composition comprises one or more surfactants.

21. The film forming composition according to claim 20, wherein the surfactant is selected from the group comprising sodium lauryl sulphate (SLS), dioctyl sodium sulfosuccinate (DSS), benzalkonium chloride, benzethonium chloride, cetrimide (trimethyl-tetradecylammonium bromide), fatty acid sugar esters, glyceryl monooleate, polyoxyethylene sorbitan fatty acid esters, polyvinyl alcohol, dimethylpolysiloxan, sorbitan esters or lecithin.
22. The film forming composition according to any one of claims 20 or 21, wherein the content of surfactant is 0.01 to 3% by weight related to the amount of pullulan.
23. Container for unit dosage forms for agrochemicals, seeds, herbs, foodstuffs, dyestuffs, pharmaceuticals, or flavouring agents produced from the film forming composition according to any one of claims 1 to 22.
24. Container according to claim 23 which is a capsule, preferably a pharmaceutical capsule.
25. Container according to claim 23 or 24, wherein the container comprises a coating.
26. Container according to claim 25, wherein the coating is selected from the group comprising cellulose acetate phthalate, polyvinyl acetate phthalate, methacrylic acid gelatines, hypromellose phthalate, hydroxypropylmethyl cellulose phthalate hydroxyalkyl methyl cellulose phthalates, hydroxypropyl methylcellulose acetate succinate or mixtures thereof.
27. Container according to claim 25, wherein the coating is a surfactant.

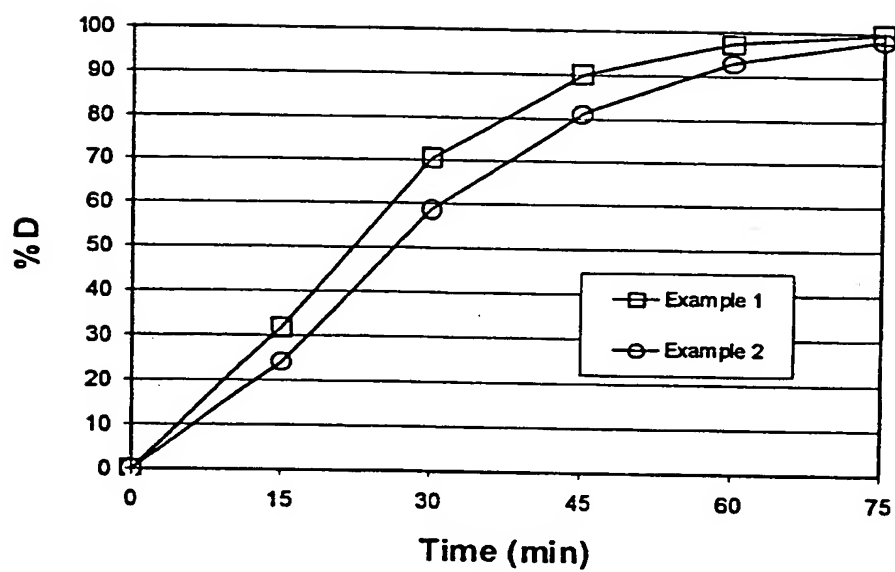


28. Container according to claim 27, wherein the coating is in the range of 0.5 to 100 microns.
29. Container according to claims 27 or 28, wherein the surfactant is selected from the group comprising sodium lauryl sulphate (SLS), dioctyl sodium sulfosuccinate (DSS), benzalkonium chloride, benzethonium chloride, cetrimide (trimethyltetradecylammonium bromide), fatty acid sugar esters, glyceryl monooleate, polyoxyethylene sorbitan fatty acid esters, polyvinyl alcohol, dimethylpolysiloxan, sorbitan esters or lecithin.
30. A caplet encapsulated in a film forming composition according to one of claims 1 to 21.
31. Container comprising two halves forming a capsule, wherein the the container is sealed with one or more layers of the composition according to claims 1 to 22.
32. Container according to claim 31 wherein the capsule halves are sealed by a liquid fusion process.
33. Container according to claim 30 or 32, wherein the capsule is a container according to one of claims 23 to 25.
34. Container according to any one of claims 23 to 33 wherein a product filled in the container is releasable at a low temperature such as at room temperature.
35. An aqueous solution of the film forming composition according to any one of claims 1 to 22 for the manufacturing of capsules.

36. The aqueous solutions according to claim 35, comprising pullulan in an amount of 10 to 60%, preferably 15 to 40% by weight of the aqueous solution.
- 5 37. The aqueous solution according to claim 35 or 36, comprising setting agent in an amount of 0.01 to 5%, preferably 0.03 to 1.0% by weight of the aqueous solution.
- 10 38. The aqueous solution according to any one of claims 35 to 37, further comprising cations in an amount less than 3%, preferably 0.01 to 1% by weight of the aqueous solution.
- 15 39. The aqueous solution according to any one of claims 35 to 38, further comprising sequestering agents in an amount less than 3%, preferably 0.01 to 1% by weight of the aqueous solution.
40. Use of the aqueous solution according to any one of claims 35 to 39 for the manufacturing of hard capsules in a dip moulding process.
- 20 41. Manufacturing of hard capsules from the aqueous pullulan solution according to any one of claims 35 to 39 in a dip moulding process with conventional hard gelatine capsules process parameters and equipment.

1/1

FIG. 1



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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/06843

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08J5/18 C08L5/00 A61K9/48

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08J C08L A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

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X	<p>US 4 623 394 A (SATOSHI NAKAMURA ET AL.) 18 November 1986 (1986-11-18) example 3; table I</p>	1-30

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Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

### \* Special categories of cited documents :

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Date of the actual completion of the international search

15 November 2000

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# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/EP 00/06843

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